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(54) EPOXY RESIN COMPOSITION, FILM ADHESIVE, AND ADHESIVE- COATED COPPER FOIL

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain resin composition capable of being formed into a film without using any glass fibers as a reinforcement by using an epoxy resin, a curing agent for the epoxy resin, a phenol/polybutadiene adduct, and polyvinyl butyral as the essential components.

SOLUTION: The curing agent for the epoxy resin is exemplified by dicyandiamide, an acid anhydride, or a phenolic curing agent. The phenolic curing agent is desirably a phenol novolak resin. The curing agent is usually used in an amount of 2-5 pts.wt., (for dicyandiamide) or of 30-80 pts.wt. (for a phenolic curing agent) per 100 pts.wt. epoxy resin. It is desirable that 100 pts.wt. epoxy resin is compounded with 30-100 pts.wt. phenol/polybutadiene adduct and 10-50 pts.wt. polyvinyl butyral. The composition may contain a filler, desirably, in an amount of 50-200 pts.wt. per 100 pts.wt. epoxy resin.

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## CLAIMS

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[Claim(s)]

[Claim 1]An epoxy resin composition which becomes considering an epoxy resin and its hardening agent, phenols addition polybutadiene, and a polyvinyl butyral as an essential ingredient.

[Claim 2]Film glue which makes film state the epoxy resin composition according to claim 1 and to which it comes to carry out semi-hardening.

[Claim 3]Copper foil with adhesives which is applied to one side of copper foil by using the epoxy resin composition according to claim 1 as a varnish, and comes to carry out semi-hardening.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to copper foil with an epoxy resin composition, film glue, and adhesives.

[0002]

[Description of the Prior Art]Since the cellular phone is electronic equipment treating high frequency, it is needed for the insulating layer of a printed wired board used for a cellular phone that it is a lower dielectric constant. Since the hardened material is a lower dielectric constant, the epoxy resin composition which becomes considering an epoxy resin, its hardening agent, and phenols addition polybutadiene as an essential ingredient fits such a use.

[0003]The small weight saving of the cellular phone is carried out increasingly in recent years, and the thin printed wired board is used. In order to slim down a printed wired board, it is necessary to slim down an insulating layer. Then, the method of forming an insulating layer and a conductive layer using the method or the copper foil with adhesives which forms an insulating layer using film glue is adopted increasingly. A laser process is increasingly adopted as various kinds of processing means, such as forming a viahole.

[0004]

[Problem(s) to be Solved by the Invention]However, in order for the epoxy resin composition which becomes as an essential ingredient to make film state an epoxy resin, its hardening agent, and phenols addition polybutadiene, it was required although glass fiber was added as a reinforcing member. Since the melting point of glass fiber is high, laser beam machining is difficult for the insulating layer which contains glass fiber as a reinforcing member.

[0005]without the invention according to claim 1 becomes considering an epoxy resin, its hardening agent, and phenols addition polybutadiene as an essential ingredient and it adds glass fiber as a reinforcing member -- a film -- it aims at providing the epoxy resin composition [-izing / an epoxy resin composition ].

[0006]An object of the invention according to claim 2 is to provide the film glue which used the epoxy resin composition according to claim 1.

[0007]An object of the invention according to claim 3 is to provide the copper foil with adhesives which used the epoxy resin composition according to claim 1.

[0008]

[Means for Solving the Problem]The invention according to claim 1 is an epoxy resin composition which

becomes considering an epoxy resin and its hardening agent, phenols addition polybutadiene, and a polyvinyl butyral as an essential ingredient.

[0009]The invention according to claim 2 is film glue which makes film state the epoxy resin composition according to claim 1 and to which it comes to carry out semi-hardening.

[0010]The invention according to claim 3 is copper foil with adhesives which is applied to one side of copper foil by using the epoxy resin composition according to claim 1 as a varnish, and comes to carry out semi-hardening.

[0011]

[Embodiment of the Invention]As an epoxy resin, there is no restriction in particular and For example, a bisphenol system epoxy resin, A phenol novolac system epoxy resin, a bisphenol novolac system epoxy resin, An alkylphenol novolac system epoxy resin, a polyphenol system epoxy resin, a polyglycol system epoxy resin, annular aliphatic series system epoxy resins, these halogenides, these hydrogenation things, etc. can be mentioned. These may be used alone and may be used combining two or more kinds.

[0012]As a hardening agent of an epoxy resin, dicyandiamide, an acid anhydride, a phenol system hardening agent, etc. are mentioned. As a phenol system hardening agent, bisphenol A, the bisphenol F, polyvinyl phenol, phenol novolak resin (a halogenide or a hydride is included), bisphenol A novolak resin (a halogenide or a hydride is included), etc. are mentioned. Especially, since the heat resistance of a hardened material is excellent, phenol novolak resin is preferred. The blending ratio of a hardening agent is suitably chosen in the range of 30 to 80 weight section by dicyandiamide to epoxy resin 100 weight section at two to 5 weight section, and a phenol system hardening agent.

[0013]A hardening accelerator can also be blended by necessity besides a hardening agent. As a hardening accelerator, an imidazole compound, an organophosphorus compound, tertiary amine, quarternary ammonium salt, etc. are mentioned. The blending ratio of a hardening accelerator is suitably chosen in the range of 0.01 to 20 weight section to epoxy resin 100 weight section.

[0014]Phenols addition polybutadiene is obtained by making phenols add to polybutadiene. The copolymer which uses butadiene, such as a copolymer of the copolymer of butadiene and vinyl monomers, such as styrene, besides a butadiene homopolymer or butadiene, and diolefins, such as isoprene, as a polymerization component is included in the polybutadiene used here. As phenols made to add to polybutadiene, what is chosen from monohydric phenol, polyhydric phenol, or these alkylation objects can be used. As for phenols addition polybutadiene, it is preferred to blend in the range of 30 to 100 weight section to epoxy resin 100 weight section. The loadings of phenols addition polybutadiene are in the tendency for a dielectric constant to become it high that they are less than 30 weight sections to epoxy resin 100 weight section, It is more preferred to be in the tendency for heat resistance to fall, when 100 weight sections are exceeded, and to blend phenols addition polybutadiene in the range of 50 to 80 weight section to epoxy resin 100 weight section.

[0015]A polyvinyl butyral is resin which makes polyvinyl alcohol and butylaldehyde react by an acid catalyst, and generates them, and it is known that compatibility with other resin is good. In this invention, the thing of the degree % and the degrees of polymerization 1500-2500 of 60-90 mol of butyral-izing is used preferably. There is a tendency for the degree of butyral-izing to become insufficient [ heat resistance ] for it to be less than [ 60 mol % ], and when 90-mol % was exceeded and it is considered as a varnish, the tendency for viscosity to become high is shown. A degree of polymerization tends to become

insufficient [ heat resistance ] for it to be less than 1500, and when 2500 was exceeded and it is considered as a varnish, the tendency for viscosity to become high is shown. From this, the thing of the degree % and the degrees of polymerization 2000-2400 of 70-85 mol of butyral-izing is used more preferably. A commercial item can be used as such a polyvinyl butyral. As a commercial item of a polyvinyl butyral, S REXX BX-1, S REXX BX-2, S REXX BX-5 (above Sekisui Chemical Co., Ltd. trade name), The DENKA butyral 5000A, the DENKA butyral 6000C, DENKA butyral 6000EP (above DENKI KAGAKU KOGYO K.K. trade name), etc. are mentioned. As for a polyvinyl butyral, it is preferred to blend in the range of ten to 50 weight section to epoxy resin 100 weight section. When the loadings of a polyvinyl butyral film-ize that they are less than ten weight sections to epoxy resin 100 weight section, are in the tendency which becomes weak and exceed 50 weight sections, it is in the tendency for heat resistance to fall. It is more preferred to blend a polyvinyl butyral from this in the range of 20 to 40 weight section to epoxy resin 100 weight section.

[0016]Like a common epoxy resin composition, in order to improve increase in quantity and a mechanical property, a filler may be blended. It is preferred to use silica as a filler to blend, since a dielectric constant is not influenced. The silica used will not have restriction, especially if blended with electric insulation resin. Loadings are suitably chosen at a rate of 50 to 200 weight section to epoxy resin 100 weight section.

[0017]Future processes are presented with the epoxy resin composition which becomes this invention as an adhesives varnish. As a solvent of an adhesives varnish, acetone, methyl ethyl ketone, toluene, Xylene, methyl isobutyl ketone, ethyl acetate, ethylene glycol monoethyl ether, ethylene glycol monomethyl ether, methanol, ethanol, N,N-dimethylformamide, N,N-dimethylacetamide, etc. can be used. These may be used alone and may be used combining two or more kinds. The blending ratio of a solvent has the preferred range of one to 200 weight section to solid content 100 weight section of an epoxy resin composition, and its range of 30 to 100 weight section is more preferred. It is in the tendency for coating nature to worsen that it is less than one weight section, and when 200 weight sections are exceeded, it is in the tendency for the heat resistance after shaping to worsen.

[0018]Carry out coating of the adhesives varnish prepared as mentioned above to one side of a carrier film, heat it, and a solvent is removed, and the adhesive sheet of this invention is prepared by making thermosetting resin into a semi hardened state. As a coating method, it can be based, for example on publicly known methods, such as the knife coating-machine method and the casting method, and there is no restriction in particular. The thickness of a sheet is suitably chosen according to a use in 30-150 micrometers. It is in the tendency to become it difficult to secure the insulation of an inner layer circuit and an outer layer circuit that the thickness of a sheet is less than 30 micrometers. When it exceeds 150 micrometers, it is to form a sheet smoothly in the tendency which becomes difficult.

[0019]As a carrier film, metallic foil, such as copper foil and aluminum foil, polyester film, a polyimide film, a polyethylene terephthalate film, etc. can be used. Since a carrier film is usually removed, it can use what processed the surface with the release agent. When copper foil is used as a carrier film, it is useful by not removing a carrier film and using it to especially manufacture of the multilayered circuit board etc. which can form an insulating layer and a conductive layer simultaneously and have IVH.

[0020]

[Example]an example 1 bisphenol-A-type epoxy resin (the weight per epoxy equivalent 177.5 and oil recovery shell epoxy incorporated company make.) Epicoat 828 (trade name) -- use 100 weight section

and phenol addition polybutadiene (60-% of the weight solution (solvent: -- xylene / methyl-ethyl-ketone partially aromatic solvent.)) the mixture ratio (weight ratio) -- xylene/methyl-ethyl-ketone =10/30, and the Nippon Oil chemicals incorporated company make. PB-1000 (trade name) -- use 125 weight section and a polyvinyl butyral (degree % of 70 mol of butyral-izing.) Methyl-ethyl-ketone 60 weight section was made to carry out dissolution distribution of use 30 weight section and the silica 125 weight section the degree of polymerization 2000, made in Denker, Inc., and for the DENKA butyral 5000A (trade name), and the adhesives varnish was prepared.

[0021]the prepared adhesives varnish -- a 50-micrometer-thick polyethylene terephthalate film (the Toray Industries, Inc. make.) Using use as a carrier film, coating of the lumiler (trade name) was carried out with the conventional method so that the thickness after semi-hardening might be set to 80 micrometers, and semi-hardening was carried out by heating for 10 minutes at 150 more \*\* for 5 minutes at 130 \*\* continuously for 10 minutes by 60 \*\*. Next, when the polyethylene terephthalate film was removed, film state was able to be maintained also in the state where there is no polyethylene terephthalate film.

[0022]Next, semi-hardening was carried out to carrying out coating of the prepared adhesives varnish to an 18-micrometer-thick copper foil roughened surface with a conventional method so that the thickness after semi-hardening may be set to 80 micrometers, and heating it to it for 10 minutes at 150 more \*\* for 5 minutes at 130 \*\* continuously for 10 minutes by 60 \*\*, and copper foil with adhesives was produced. The obtained copper foil with adhesives was piled up as two sheets and copper foil became the outside, heat pressing arrival was carried out for 90 minutes by the temperature of 175 \*\*, and pressure 3MPa, adhesives were stiffened, and the double-sided copper-clad hardened material was obtained. After removing double-sided copper foil completely by etching about the obtained double-sided copper-clad hardened material, the specific inductive capacity for which it asked with the bridge method (frequency of 1 MHz) provided in JIS C 6481 was 3.5.

[0023]Comparative example 1 polyvinyl butyral was not blended, and also the adhesives varnish was prepared like Example 1. Like Example 1, coating of the prepared adhesives varnish was carried out to the polyethylene terephthalate film, and semi-hardening was carried out to it. And when the polyethylene terephthalate film was removed, it cracked and became unusable. Next, coating of the prepared adhesives varnish was carried out to the 18-micrometer-thick copper foil roughened surface, and it was made to carry out semi-hardening like Example 1. The obtained copper foil with adhesives was piled up as two sheets and copper foil became the outside, adhesives were stiffened like Example 1, and the double-sided copper-clad hardened material was obtained. After removing double-sided copper foil completely by etching about the obtained double-sided copper-clad hardened material, the specific inductive capacity for which it asked with the bridge method (frequency of 1 MHz) provided in JIS C 6481 was 3.9.

[0024]The copper foil with adhesives produced in Example 1 was put on the glass cloth base material epoxy resin copper clad laminate which performed and roughened the oxidation reduction process, and by the temperature of 175 \*\*, and pressure 3MPa, heat pressing was carried out for 90 minutes, and it pasted together. The copper foil side pasted together was etched and a conformal mask 100 micrometers in diameter was produced. And with the carbon-dioxide-gas-laser processing machine (Hitachi elaborate incorporated company make and LCO-1A21 (trade name) are used), when laser beam machining was performed with the processing frequency of 500 Hertz, and 10 microseconds of pulse width, good IVH was able to be formed by three exposures.

[0025]

[Effect of the Invention] Film-izing is possible for the epoxy resin composition which becomes this invention, without adding glass fiber as a reinforcing member. And laser beam machining of a hardened material becomes easy by using the copper foil with film glue or adhesives which becomes this invention.

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